

Li⁺ ion emission from a hot-plate alumina-silicate source stimulated by flash heating with an infrared laser

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The Neutralized Drift Compression Experiment-II accelerator under construction at Lawrence Berkeley National Laboratory has been designed to employ a lithium-doped alumino-silicate (Al-Si) hot-plate surface-ionization ion source. In order to achieve the design 1 mA/cm² current density, the emitter must be constantly kept at a high temperature, leading to the accelerated loss of Li material as ions or neutrals. As a result, the estimated lifetime of the source is 50 h. This lifetime can be extended if the source is kept at low temperature during standby, and pulse heated to the high temperature during the ion extraction phase only. A pulsed heating technique based on an infrared laser (CO₂ gas discharge, $\lambda = 10.6 \mu\text{m}$) is described in this paper. The feasibility of ion current emission stimulated by flash heating with an infrared (IR) laser was demonstrated. High repeatability of the laser-stimulated ion current was observed, creating an opportunity for modulation and gating of the ion current with a laser pulse. It was found that with the available low power ($\approx 115 \text{ W/cm}^2$) IR laser, current densities as high as 0.8 mA/cm² could be achieved with a 2.8 mm diameter source. Various approaches for scaling to a larger (10 cm diameter) source and the application of short pulse, high power lasers are discussed. The results and conclusions of this paper may apply to various species of hot-plate ion sources. © 2011 American Institute of Physics. [doi:10.1063/1.3555334]

I. INTRODUCTION

The work presented in this paper is a part of the lithium hot-plate source development for the Neutralized Drift Compression Experiment-II (NDCX-II) (Refs. 1 and 2) accelerator at Lawrence Berkeley National Laboratory (LBNL). The primary purpose of NDCX-II is to carry out laboratory plasma and astrophysics high-energy-density LABORATORY plasma experiments (HEDLP) with intense ion beams. One aim of these experiments is to investigate the properties of matter under extreme conditions, for example, to study the equation of states of various metals near a critical point, which is the least studied region of the phase diagram.³ Another aim is to carry out experiments relevant to the physics of inertial fusion energy targets driven by beams of heavy ions.

The output kinetic energy of NDCX-II will initially be 1.2–1.5 MeV.¹ For a several micrometer thin foil, Li⁺ ions at this energy are used in order to match the desired penetration range and achieve uniform volumetric heating. The ion beam requirements for heavy-ion fusion and HEDLP are related. In both cases, a low duty factor ion beam pulse must be transversely focused to a submillimeter spot and longitudinally compressed to a nanosecond pulse. The ion beam must be launched from an ion source that simultaneously offers high current, high brightness, and stable on–off gating control with fast rise time. The requirements for near-term experiments have led to the selection of the hot-plate surface ionization type of ion source, a lithium-doped alumino-silicate (Al-Si) source similar to the potassium source currently used in NDCX-I.⁴

Due to the high ionization potential of lithium, it is generally more difficult to make Li⁺ than the other alkaline ions (e.g., K⁺, Na⁺, and Cs⁺).⁵ The goal of the NDCX-II ion

source is to provide about 100 mA of Li⁺ beam current for $<1 \mu\text{s}$ with the highest possible current density limited by the operating temperature of the hot plate (tungsten substrate).¹ The total charge the accelerator must deliver is of order 50 nC. Additional NDCX-II requirements include ion pulses fired at a 2 pulses/min repetition rate, and sufficiently uniform emission that the beam emittance is not significantly degraded, ideally achieved via operation in a mode wherein the entire emitting surface is in the space-charge-limited regime.

Recent test data⁶ have shown that emission at a 2 mA/cm² current density would require an emitter temperature exceeding $\approx 1300^\circ\text{C}$, which is near the melting temperature of the alumino-silicate material. Furthermore, the data suggest that a current density of 1 mA/cm² is a good compromise between operating temperature, lifetime, and stable emission. Based on the observations to date, it has been found that for an emitter temperature of $\approx 1270^\circ\text{C}$, an injector designed for a space charge limited Li source of $J \sim 1 \text{ mA/cm}^2$ will have a lifetime of order 50 h, enabling approximately 1 week of running on NDCX-II. This short lifetime of the source is an issue since hundreds of operational hours are needed for tuning the accelerator and carrying out target experiments. For comparison, the lifetime of the NDCX-I potassium source, which operates at 1050°C , is hundreds of hours.⁴

The large discrepancy in lifetimes is due to the fact that lithium has a higher ionization potential (5.39 eV) than potassium (4.34 eV), and according to the Saha-Langmuir equation this results in a lower number of emitted Li⁺.⁵ Thus, in order to achieve higher Li⁺ current, temperature must be increased, which has a negative side, since it is accompanied by an increase in loss rate of lithium from the alumino-silicate material (either as ions or as neutral atoms), hence shorter lifetime.

One strategy to extend the lifetime is to keep the source at a lower temperature and only raise it to high temperature during extraction. In this mode, fewer lithium atoms are lost between shots and the lifetime is increased significantly. There are various methods of pulse heating the source, e.g., incandescent heating of the metallic source substrate, electron bombardment heating, RF microwave-heating, and flash heating with photons. Factors including efficiency, heater complexity, availability of space in an injector, cost, reliability, repeatability, and scalability to large source diameter must be considered.

In this paper, we investigate an approach based on flash heating the ion source using a laser. A laser-based strategy has several advantages. Industry has a broad range of powerful, easy-to-maintain lasers which are commercially available. The pulse timing and format can be controlled reliably and more importantly, the laser energy can be pointed precisely, allowing for the direct surface heating of only the source material. For comparison, in the case of incandescent heating, the source material is heated indirectly through thermal contact with the hot metal substrate. Moreover, a laser spot on a source can be focused to various shapes, including super-Gaussian (flat distribution), elliptical, line, grid, etc.

The choice of light wavelength is critical for flash heating because the photons must be absorbed in the Al-Si. Otherwise, the energy is transferred to the metal substrate and the source is heated indirectly through thermal exchange with the substrate, in a process similar to incandescent heating. The spectral transmission of alumina-silicate material is not known; however, due to its glassy appearance, it is fair to estimate that it is similar to fused silica glass, which is opaque for ultraviolet (UV) and infrared (IR) wavelengths and transparent for visible and near-infrared light. Thus, one should either use a UV or an IR light source. In this paper, the ap-

plicability of intense IR light for the flash heating of Al-Si is investigated.

II. DESCRIPTION OF EXPERIMENT AND METHODS

Experiments were carried out with a scaled down lithium doped alumina-silicate source. The preparation of the source was performed in the same vacuum chamber ($P = 10^{-6}$ Torr) used to carry out the flash-heating experiment. Al-Si powder was placed in the dimple of a thin molybdenum (Mo) strip that could be directly heated to melt the Al-Si (exceed 1350°C) by passing electrical current through the strip.⁶ The solidified material produced a 2.8 mm diameter, 30 mg source. An electrode, later referred to as the cathode, with a negative voltage bias can be moved into position directly above the dimple to collect current, thus forming a low voltage dc ion diode to make an *in situ* ion emission measurement.

A layout of the experiment is presented in Fig. 1. A CO₂ gas laser, Synrad model 48-2 with 25 W of continuous wave (CW) output at $\lambda = 10.6\ \mu\text{m}$, was used as the pulse heater. The unfocused laser beam (5 mm diameter, 4.0 mR divergence) was directed onto the source by a pair of gold mirrors through a Zinc-Selenium vacuum window (95% transmission at $10.6\ \mu\text{m}$). The CW output can be gated down to a minimum of 100 μs duration with a 10 KHz repetition frequency. The laser and a pyrometer used for dynamic temperature measurement had a clear angular view of the source through machined holes in the cathode plate. The cathode was a 1 mm thick Mo plate, biased to $-150\ \text{V}$, and positioned 30 mm away from the source. In this setup, it takes approximately 1 μs for a Li^+ ion to traverse the diode gap. Current was measured through a 100 k Ω resistor connected in series with the ion diode. Voltage across the resistor was measured with a differential voltage probe with an attenuation factor of 50. Thermionic

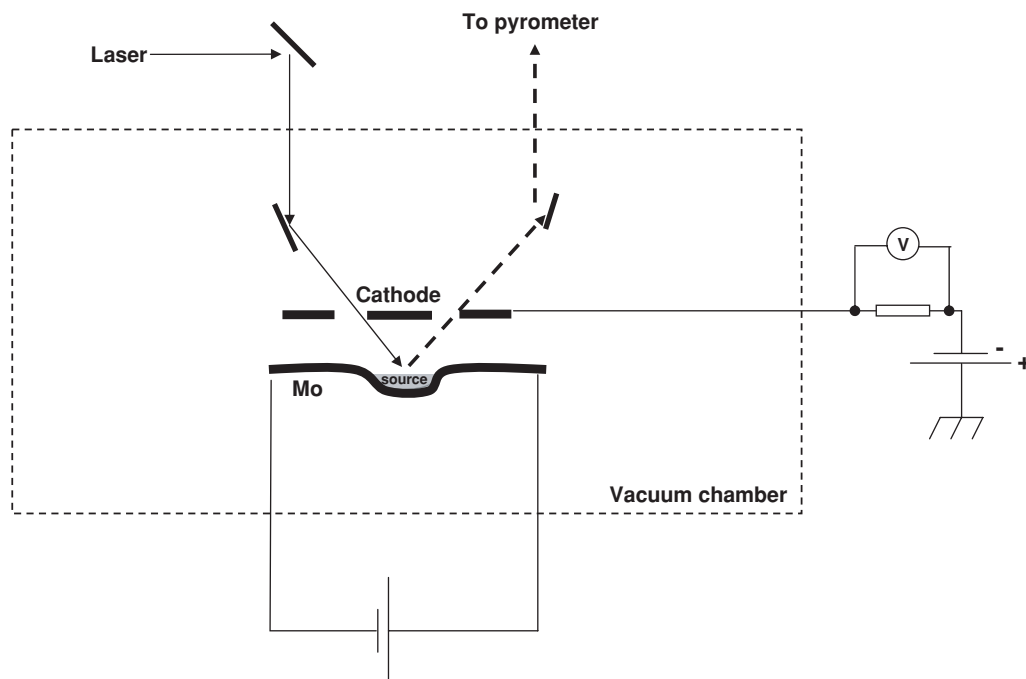


FIG. 1. Scheme of the experiment (side view).

electrons, emitted by Mo at high temperature, were suppressed by the negative biasing of the cathode.

First, we verified that the Al-Si absorbs the $\lambda = 10.6 \mu\text{m}$ light. To do this, we irradiated a 1 mm thick free-standing Al-Si flake (no substrate) and confirmed that most of the laser light was absorbed in the material. There were no available means to determine the depth of the light penetration. In addition, it was measured that the most of the IR light is reflected by Mo and thus the filament cannot be heated by the laser. This is relevant since the beam was not focused and both Al-Si and Mo were illuminated. Considering that only a 2.8 mm diameter area of Al-Si was absorbing energy, the effective laser power after the correction for effective area is 7 W (114 W/cm^2 power density). It was also assumed that there was a 10% combined photon loss at the vacuum window and guiding mirrors.

The temperature of the Al-Si was measured by a custom one-channel, 700 nm (FWHM 25 nm) optical pyrometer (2 mm probing spot diameter, 2.5 ns temporal response, and $\approx 750^\circ\text{C}$ lowest detectable temperature). A single-wavelength pyrometer measures the brightness (black body) temperature, assuming the emissivity is equal to one. However, the emissivity of real bodies, including Al-Si, is less than one and consequently, a single-wavelength pyrometer produce temperature values lower than the true temperature.⁷ The term “temperature,” as used in this paper, represents the brightness of a surface expressed in degrees celsius and should be considered a relative measure rather than an absolute one.

III. EXPERIMENTAL RESULTS

The dependence of Li^+ current density (J) on temperature (T) was obtained in earlier incandescent heating experiments.⁶ Before proceeding with the laser heating tests, these earlier data were verified by varying the temperature of the filament (by adjusting the dc current) and measuring the corresponding ion current. Agreement was obtained, thus establishing that the measured current is indeed due to extracted Li^+ ions. According to the measured $J(T)$ function (diamonds in Fig. 8, as described below), ion current appears at $T_0 \approx 850^\circ\text{C}$ and reaches the design current density $J_d = 1 \text{ mA/cm}^2$ at $T_d = 1275^\circ\text{C}$. An ideal scenario, if sufficient laser power is available, would be to have the source heated to T_d from room temperature. In order to achieve J_d , in the case where limited laser power is available, the source must be preheated by a dc current (ideally to $T_0 \leq 900^\circ\text{C}$).

In the first series of experiments, the pulse duration of the laser (100 μs to 20 s) and the initial source temperature were varied. It was found that for the given 114 W/cm^2 laser power density, notable temperature, and current signals were occurring for pulse durations exceeding a fraction of a second. Results for an initially cold source which was heated with a 20 s pulse are shown in Fig. 2. Results for a source which was preheated to the ion emission threshold temperature are shown in Fig. 3. It can clearly be seen that ion current increases with temperature and gradually declines to zero after the laser has been shut off. Note, that due to the nonlinear behavior of $J(T)$, the current decreases much faster than the

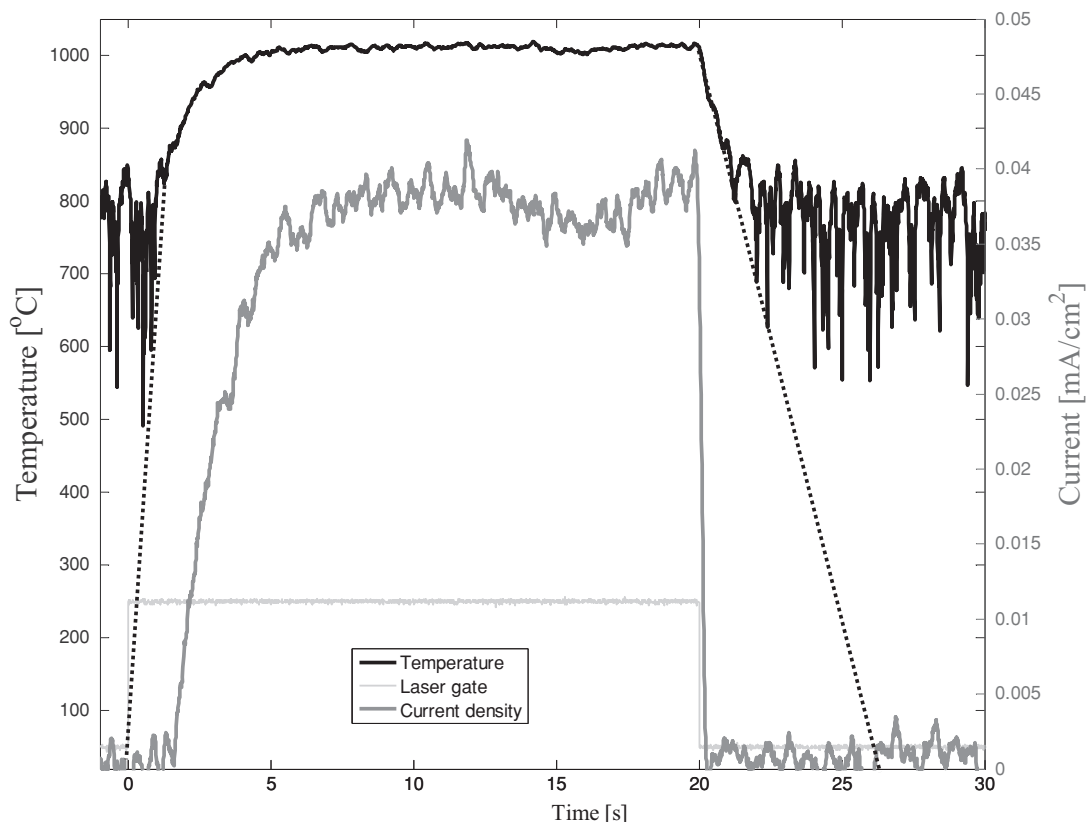


FIG. 2. Current density and temperature of the Li source (initially at room temperature) heated by a 20 s laser pulse. Pyrometer can sense temperature starting from 750°C , the low detection threshold is represented by the noise level; dashed lines are extrapolated temperature behavior.

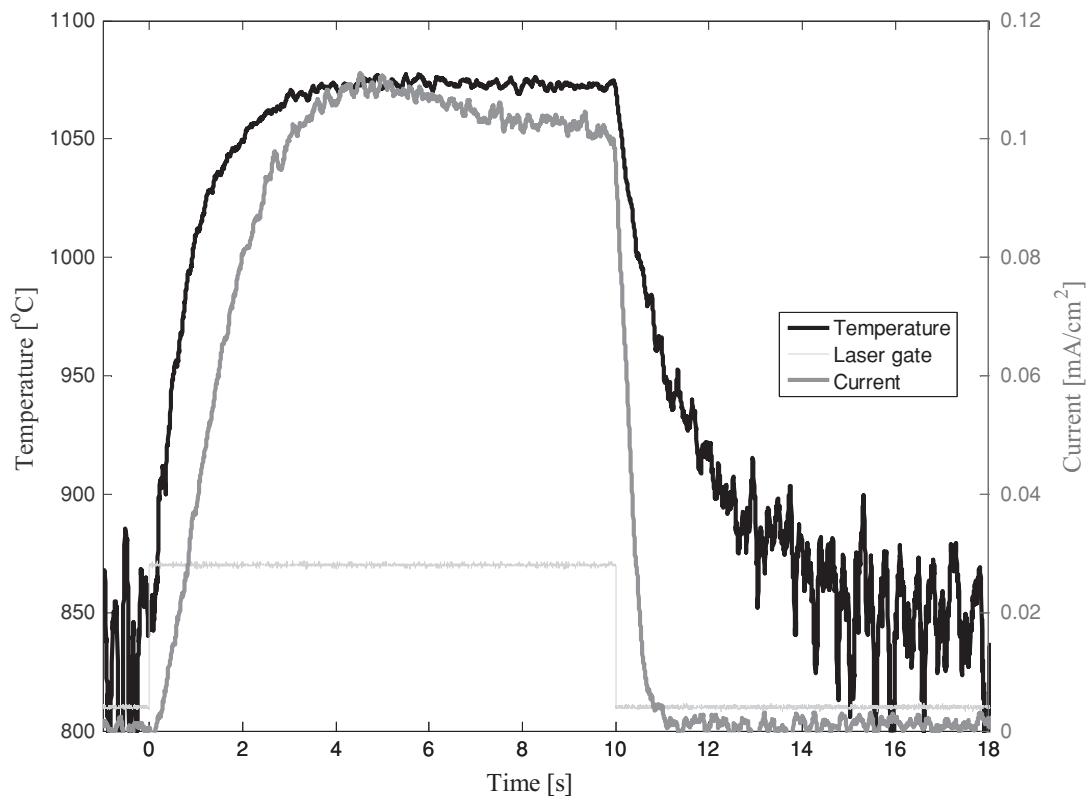


FIG. 3. Current density and temperature of the Li source heated by a 10 s laser pulse. The source was preheated to the threshold zero emission temperature (850 °C).

temperature and at milliseconds time scales it is fair to assume that the current is off when the laser is off. These findings have two important consequences. First, it demonstrates that an IR laser can be utilized for stimulation of Li^+ emission, and second, it gives the possibility of using a laser pulse as a stable on-off gating control for ion current.

In both plots, the temperature peaks after ~ 2 s and remains flat even with the laser still on. This plateau behavior indicates thermal equilibrium between the power supplied by the laser and the losses caused by heat conduction out through the Mo substrate and surface radiation. Once the plateau is reached, the laser power does not contribute to further temperature growth and is spent to maintain the constant ion current. Submicrosecond pulses are required for NDCX-II, thus the laser should be turned on for the time necessary to reach a peak temperature, i.e., ~ 2 s in our case.

The time it takes to achieve the equilibrium and the value of the equilibrium temperature depends on multiple factors including the laser power, initial temperature, geometry of the experiments, thermal properties of Mo substrate, and thermal isolation. Clearly, the 7 W heating rate is too slow, since on these long time scales relative to heat-transfer times, the thermal losses equilibrate to the input laser power before the J_d value is reached. Nevertheless, the data in Figs. 2 and 3 are an important result because it serves as a starting point for further development of the concept, which, for example, would include a more powerful laser and an optimized substrate design with minimized thermal losses.

Due to the repeated cooling and heating cycles, the surface condition (e.g., roughness) of the source changes. Surprisingly, a high degree of reproducibility of $J(T)$ was ob-

served, even when taking data several days later after multiple cycles of heating and cooling (in some cases, to room temperature). This repeatability (i.e., independence on surface condition) is an indication that the laser light is absorbed at distances larger than the typical scale size of surface defects. Since surface defects are usually on a micrometer scale, it is fair to assume that the IR light absorption depth is of the order of micrometers.

As the source cools down, the current falls to its initial value within several seconds. Considering this fact and the high degree of repeatability, it is possible to modulate (or gate) the ion current with high precision at several Hz frequency. Examples of repeatable current modulation are shown at 0.2 Hz in Fig. 4 and at 2 Hz in Fig. 5. In the example shown in Fig. 5, the time between pulses is shorter than the relaxation time (to initial temperature) resulting in the increase and equilibration to a higher average temperature. These findings make the laser flash heating suitable for a highly reproducible injector, since the laser pulse can be precisely synchronized with the ion extraction voltage.

It was possible to access higher temperatures with the modest available laser power by further increasing of the initial temperature T_0 . Current densities and temperatures recorded in this series of experiments are shown in Figs. 6 and 7. Summarized, a peak current density of $J = 0.8 \text{ mA/cm}^2$ corresponding to $T = 1175 \text{ °C}$ was achieved for initial values $T_0 = 1080 \text{ °C}$ and $J = 0.1 \text{ mA/cm}^2$. The initial temperature was not increased further due to the risk of damaging the guiding mirror and melting the source. If not for this technical limitation, the target of 1 mA/cm^2 and higher could have been easily achieved. It is worth

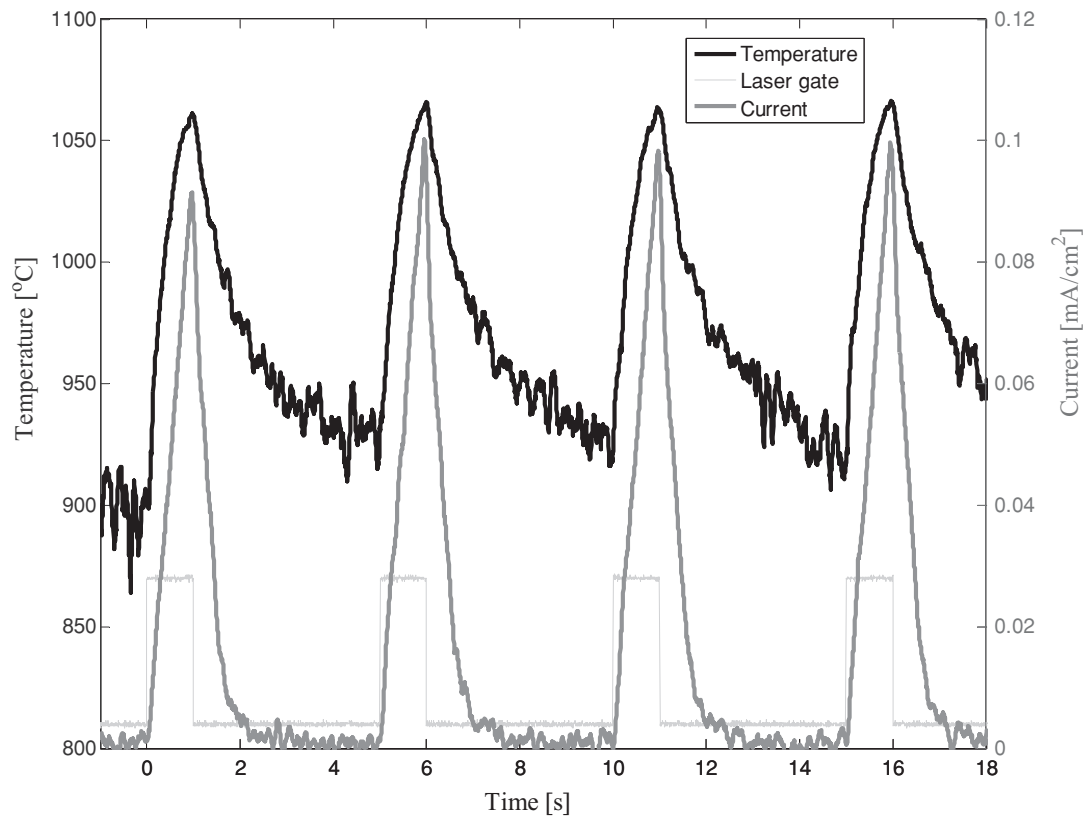


FIG. 4. Ion current and temperature modulated by a train of 1 s laser pulses fired at 0.2 Hz. Source was preheated to $T_0 = 900^\circ\text{C}$.

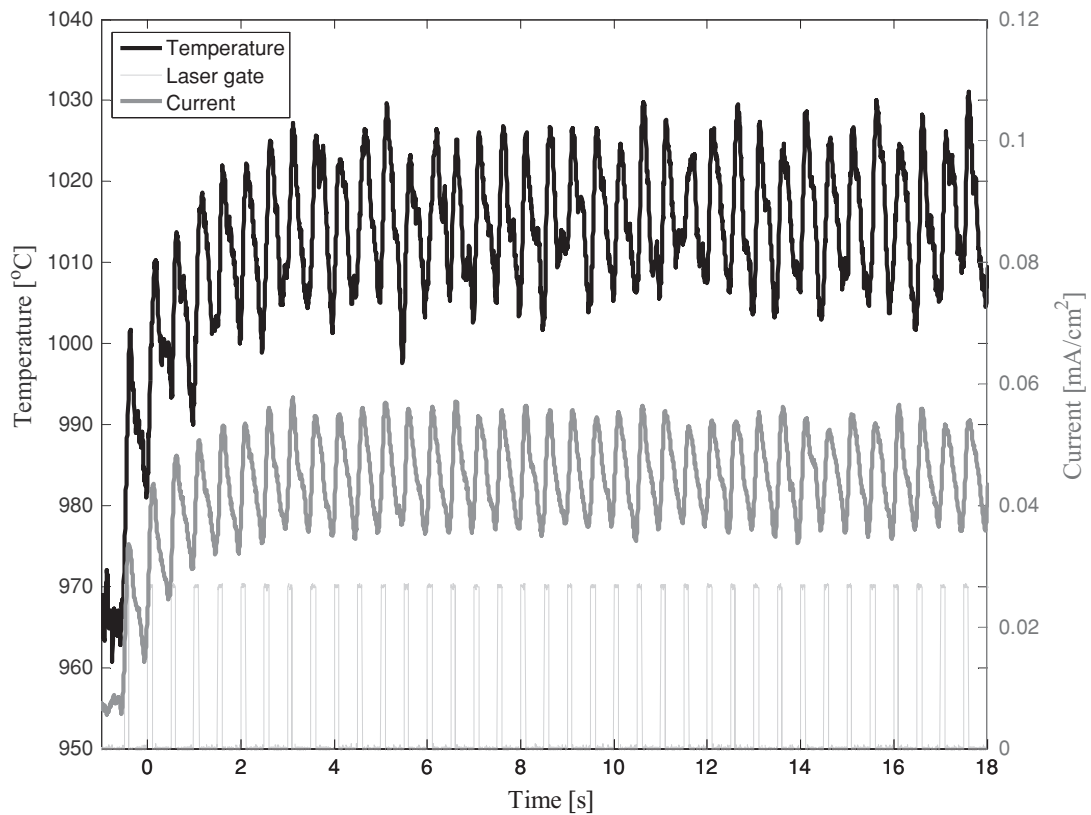


FIG. 5. Ion current and temperature modulated by a train of 100 ms second laser pulses fired at 2 Hz. Source was preheated to $T_0 = 965^\circ\text{C}$.

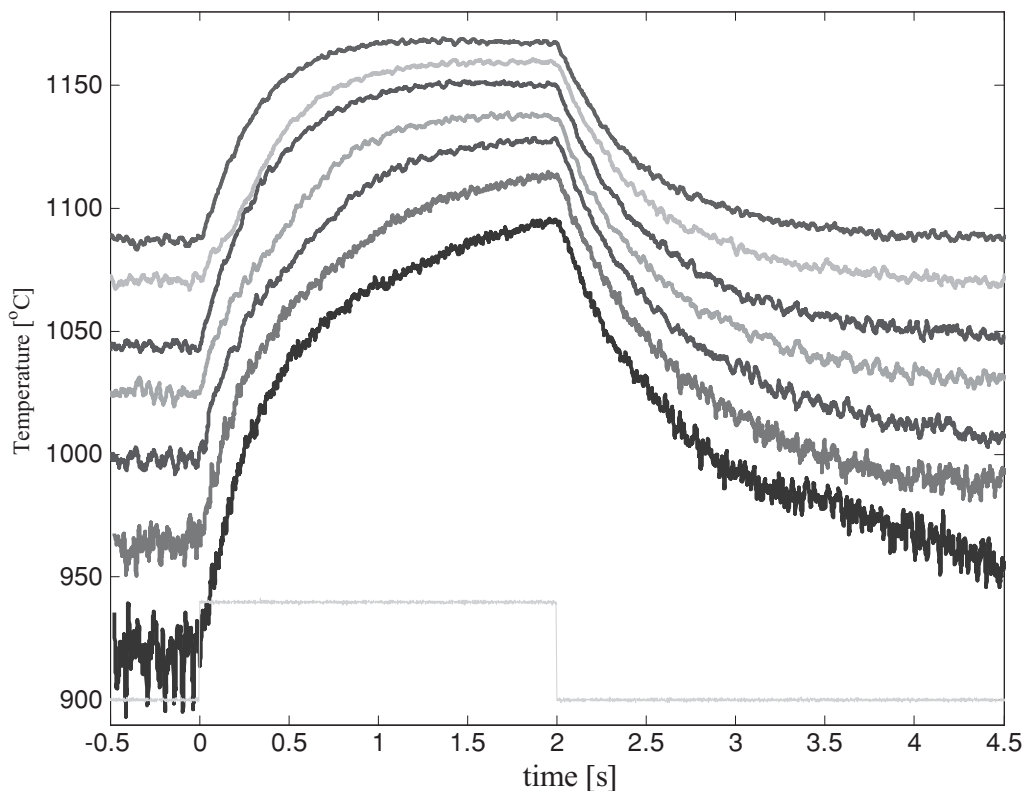


FIG. 6. A series of temperature records with increasing initial temperature.

emphasizing that the short-duration increase in current density ($0.1\text{--}0.8\text{ mA/cm}^2$) induced by this laser pulsing technique could potentially extend the lifetime of this particular 2.8 mm diameter Li source several-fold compared to a conventional dc heating approach where the source would be kept continuously at $T = 1175^\circ\text{C}$. Experimental studies of life extension will be addressed in future work.

In Fig. 7, it can be seen that, at some point, the current falls even though the temperature continues to rise or stays constant. The higher the temperature, the more pronounced is the current drop. This depletionlike behavior manifests itself in hysteresis when the data are plotted in J - T coordinates (Fig. 8). When comparing static (wherein the temperature was varied incrementally and the current was measured when thermal equilibrium had been reached in several minutes) and pulsed methods in this format, it is clear that the pulsed heating generates the same current at a lower temperature. Practically, this suggests that with this laser pulsing technique, 1 mA/cm^2 can be reached at $\sim 1175^\circ\text{C}$ (based on extrapolation), while conventional dc heating would require 1275°C . It is also important that current depletion occurs on time scales long compared to the submicrosecond NDCX-II extraction pulses, thus enabling an extraction of current at its peak before the current depletion occurs.

IV. DISCUSSION

The nature of the hysteresislike behavior is not clearly understood. Plausibly, the fact that the effect manifests itself in pulsed experiments only is an indication of diffusion dynamics of Li atoms within the source. dc heating re-

sults in an equilibrated outflow of Li^+ ions which are continuously resupplied from the bulk of the Al-Si. Since ions have a finite diffusion time, the concentration of ions at the surface is lower than that in the bulk of the Al-Si. When the surface is flash heated, the ion concentration has not reached the lower equilibrium value, resulting in initially high currents.

From a practical point of view, it is important to investigate the applicability of laser flash heating to a larger diameter source. From the simplest experiments described in this paper, it was learned that 7 W of $10.6\text{ }\mu\text{m}$ radiation can heat the 2.8 mm diameter Al-Si source from 850°C (zero-emission threshold temperature) to 1080°C ($\Delta T = 230^\circ\text{C}$) in 2 s after which equilibrium is reached (Fig. 3). Thus, in the simplest linear approximation, the source temperature can be raised to the baseline 1275°C if the laser power is $\sim 13\text{ W}$, or 26 J total energy. If the latter numbers are scaled to the 10 cm diameter source, then the necessary power is 17 kW or 33 kJ total energy. Note that the power requirements might be lower if one considers the hysteresis phenomena shown in Fig. 8. The highest CW power of commercially available CO_2 lasers is 20 kW; hence, it is possible to pulse heat a 10 cm diameter source using a CW IR laser approach although this route is costly and has modest efficiency, comparable with that of straightforward incandescent heating.

If one uses a short pulse high power laser system (pulses less than a microsecond), the efficiency can be improved significantly. Li^+ ions are emitted from a several-micrometer-thick near-surface layer, and thus an economical method would be to utilize all the laser energy for heating only the

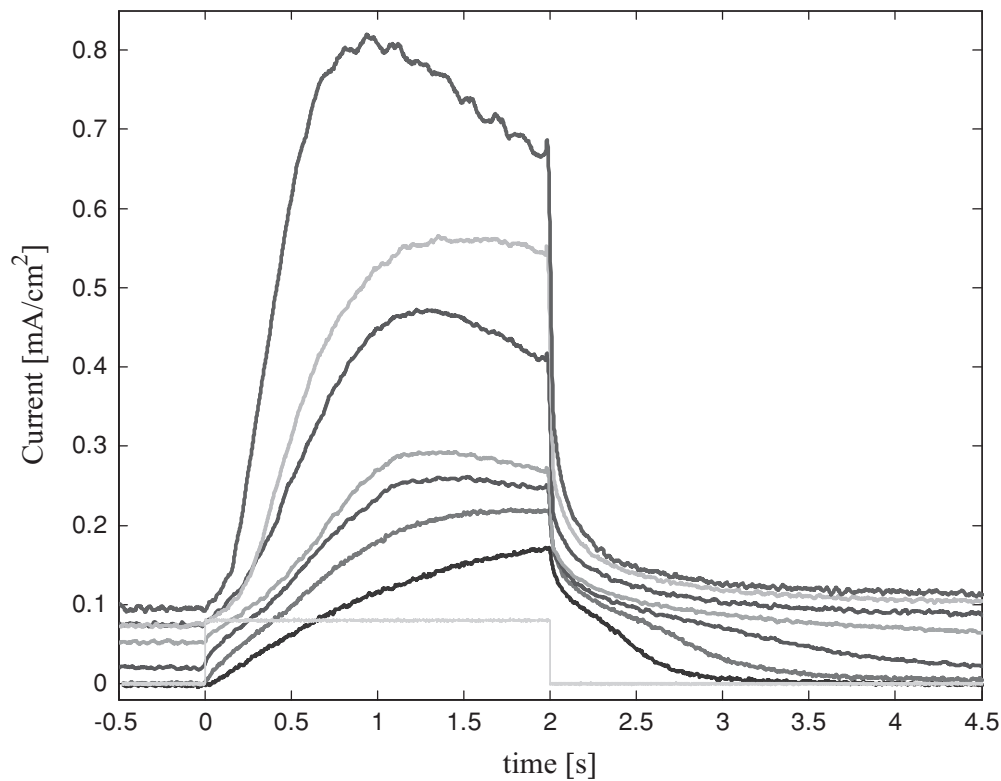


FIG. 7. Ion current density corresponding to temperature records, shown in Fig. 6.

ion-emitting layer.^{5,6} The laser energy is also absorbed in a thin near-surface layer of the thickness comparable to a fraction of the laser wavelength. The penetration depth of $10.6\ \mu\text{m}$ photons in Al-Si is not known exactly, but for the sake of simplicity, it may be assumed to be equal to one wave-

length, i.e., $d = 10\ \mu\text{m}$. As can be seen, an IR laser is an optimal choice since its photon penetration depth is comparable with the thickness of ion-emitting layer.

In addition, the efficiency can be improved further if the total mass of the heated source is reduced by using a smaller

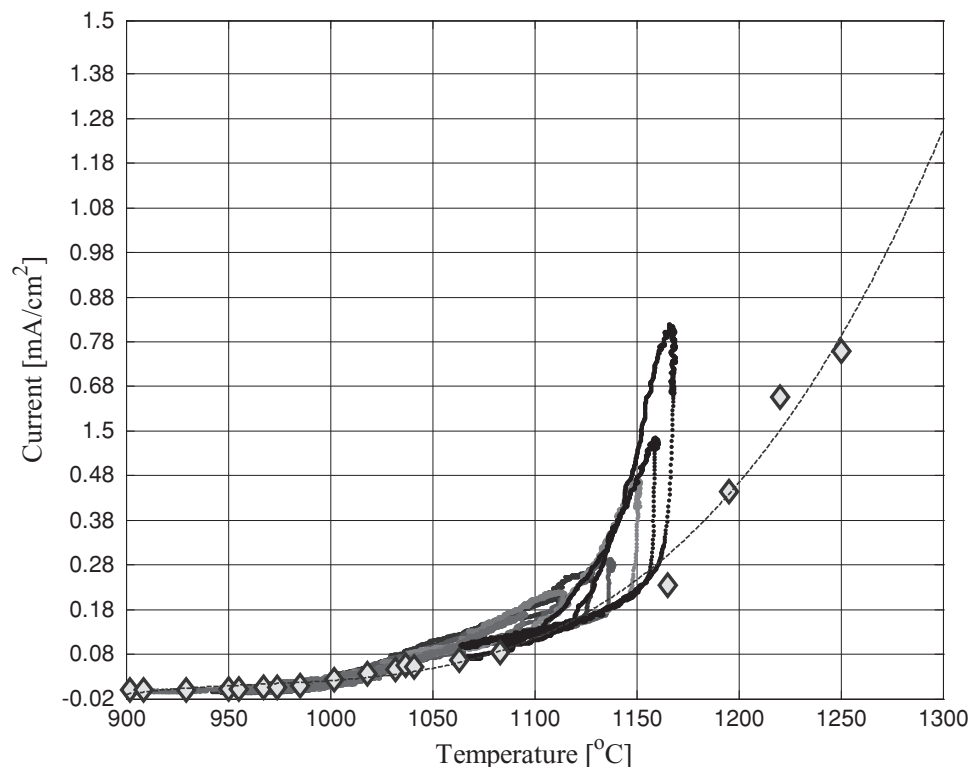


FIG. 8. Current density vs temperature in static (diamonds) and dynamic laser heating of the source. Dynamic laser heating data points represent multiple shots with various initial conditions and pulse durations.

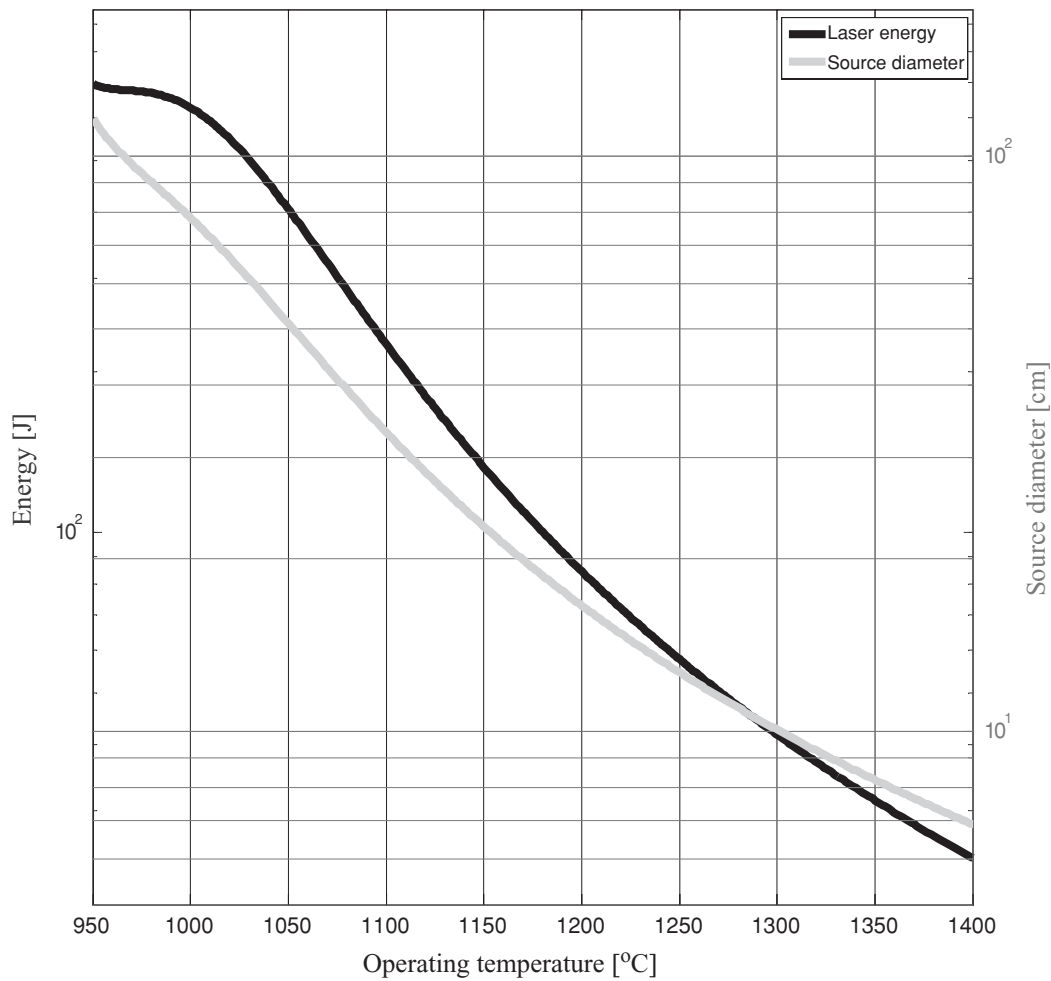


FIG. 9. Energy required to heat source to a given operating temperature (dark curve) and the corresponding source diameter necessary to produce 100 mA of Li^+ .

diameter source operated at higher temperature so that the total current is unchanged. Possible combinations of source size and operating temperature are governed by the simple expression,

$$100 \text{ mA} = J(T) \cdot \pi \left(\frac{d}{2} \right)^2. \quad (1)$$

Here $J(T)$ is the current density function (extrapolation line in Fig. 8), d is the diameter of the source, and 100 mA is the total current required from the NDCX-II injector.

The NDCX-II injector design requires a steady 100 mA current maintained for 500 ns, which means that the operating temperature must also stay flat during the same period. Heat transfer and radiation losses for a several micrometer thick ion-emission layer are negligible on such a short time scale. This means that if a laser can deliver its energy rapidly enough, the peak temperature will stay flat since heat is not transferred to the surroundings. Thus, assuming no losses, the energy Q required to heat the surface from $T_0 = 850^\circ\text{C}$ (correspond to the ion emission threshold temperature) to T can be approximated as

$$Q(T) \approx mc(T - T_0) = \rho \cdot \delta \cdot \pi \left(\frac{d}{2} \right)^2 \cdot c(T - T_0). \quad (2)$$

Here, m is the total mass of the heated source material, $c = 0.8 \text{ J/K/g}$ is the estimated heat capacity of Al-Si (assumed to be close to that of typical glass or ceramic), $\rho = 1700 \text{ kg/m}^3$ is the density of Al-Si, and $\delta = 10 \text{ }\mu\text{m}$ is the estimated penetration depth of the IR photons. The energy requirements resulting from the solution of the above expressions are summarized in Fig. 9. The curves may seem non-intuitive, i.e., the higher the operating temperature, the lower the pulse energy and thus the smaller the source size. However, this is simply a consequence of the nearly exponential behavior of $J(T)$.

If the operating temperature equals the dc source $T_d = 1275^\circ\text{C}$ of the baseline NDCX-II scenario, then the laser pulse must contain 48 J (versus 33 kJ for the CW option). If a near-melting 1400°C operating temperature is assumed, the total energy must be 24 J and would required a 7 cm diameter source or half the area of the dc source. Note, that these pulse energy estimates can be satisfied by available laser systems. For example, several 200 ns pulse CO_2 laser systems

with pulse energies up to 12 J can be purchased at a fraction of the cost of the present NDCX-II injector.

In summary, an optimization strategy to maximize efficiency is to go to higher temperatures and current densities and to utilize smaller sources. It is worth emphasizing that the estimated values are conservative. In reality, the numbers may be more favorable since the penetration depth of IR light was likely overestimated and the high current hysteresis phenomena effect in Fig. 8, i.e., higher current at lower temperature, has not been taken into account. Nonetheless, this remains an extrapolation.

In parallel with the IR laser-based heating technique, an alternative approach which utilizes a UV laser is being investigated. An available excimer UV laser can deliver much shorter and more energetic pulses than an IR laser of comparable cost. There are several principal differences between the interaction of UV and IR photons with Al-Si which must be considered. First, the UV photons are absorbed in a nanometer thick layer, which is much smaller than the ion emitting volume. In this case, the maximum current may be restricted by the number of Li^+ ions instantaneously available in such a small volume. In addition, the absorption of this short wavelength is more sensitive to surface conditions which, in turn, affects repeatability. Second, the energy deposition mechanisms of UV and IR light in a dielectric are different. The low energy of IR photons matches the vibrational and rotational energy levels of dielectric molecules. Thus, once IR light is absorbed, it is directly transferred to thermal motion of matter. In contrast, energetic UV photons are absorbed primarily by electrons, which are excited to higher levels in the conducting band and then transfer their energy to the lattice through phonon recombination.⁸ Meanwhile, a fraction of UV photons are spent to produce photoelectrons, which do not contribute to the heating of the source. In this respect, an IR laser may be a more efficient heater since all of the energy is converted to thermal energy.

V. SUMMARY

The feasibility of ion current emission stimulated by flash heating with an IR laser has been demonstrated. The high degree of repeatability of the data demonstrates precise gating and modulation of ion current with the laser. A maximum current density of 0.8 mA/cm^2 of Li^+ ion was achieved with a 2.8 mm diameter source. The observed hysteresis of $J(T)$ is not understood and is subject to further investigation. In

a simplest approach, a costly 20 kW CW-type laser system would be necessary for the 10 cm diameter NDCX-II source. A more efficient and less expensive approach is to flash-heat the surface with a short-pulse version of a higher power CO_2 laser.

This work may lead to long-lasting alkali ion sources for accelerators used by chemical, medical, semiconductor, and nuclear industry. Lithium ions are particularly relevant for lithium battery (electrode implantation) sector, calibration of nuclear instruments, and mass spectrometry equipment. On a global scale, the hot-plate-type of sources are a primarily candidate for heavy ion fusion drivers (used for sustainable, carbon-free energy generation), where life time, efficiency, and low cost of the machine are critical issues. The work can also benefit design of ion thruster for space propulsion, where weight, compactness, and reliability are important.

ACKNOWLEDGMENTS

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